

Hall effect in PrB_6 and NdB_6

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Abstract

Hall effect was studied on the single crystals of antiferromagnets PrB_6 and NdB_6 at temperatures $2\text{K} < T < 300\text{K}$ in magnetic fields up to 8T using the sample rotation technique. At low magnetic fields $\mu_0 H \leq 1\text{T}$ Hall coefficient R_H , which is practically temperature independent in paramagnetic state at $8\text{K} \leq T \leq 70\text{K}$, is characterized by the values of $R_H(\text{PrB}_6) \sim -(4.2 \pm 0.1) \cdot 10^{-4} \text{ cm}^3/\text{C}$ and $R_H(\text{NdB}_6) \sim -(4.1 \pm 0.1) \cdot 10^{-4} \text{ cm}^3/\text{C}$. Rather different behaviour of R_H is observed in antiferromagnetic (AF) phases of these hexaborides. For PrB_6 the decrease of temperature below $T_N \approx 6.7\text{K}$ is accompanied by a noticeable ($\Delta R_H/R_H \sim 10\%$) elevation of $R_H(\mu_0 H = 1\text{T})$ to the values of $-(3.8 \pm 0.1) \cdot 10^{-4} \text{ cm}^3/\text{C}$. On the contrary, the low field Hall coefficient in NdB_6 diminishes by about 15% reaching the value $R_H \approx -(4.7 \pm 0.1) \cdot 10^{-4} \text{ cm}^3/\text{C}$ in AF state at 2.5K. The increase of magnetic field inducing magnetic transition in the commensurate magnetic phase of PrB_6 results in essential R_H changes (up to 10%) at liquid helium temperatures. The anomalous behaviour of the charge transport parameters for RB_6 ($\text{R}=\text{Pr}, \text{Nd}$) found in vicinity of Neel temperature suggests the possible effect of 5d-states spin density polarization of both in AF and paramagnetic states of the compounds under investigation.

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1.INTRODUCTION

The interest to the family of rare earth hexaborides (RB_6) is supported by their promising applications as thermoelectric and effective thermionic cathode materials [1, 2]. However, these compounds also demonstrate an exceptional variety of unusual physical phenomena. In particular, the heavy fermion compound CeB_6 was recently shown to enter into unusual AF phase with anomalous transport and magnetic properties [3]. SmB_6 is known to be an archetypal intermediate valence compound with fast charge fluctuations [4]. Europium hexaboride (EuB_6) demonstrates the colossal magnetoresistance effect in the vicinity of ferromagnetic phase transition [5]. Finally, two consecutive phase transitions - structural one induced by cooperative Jahn-Teller effect with changing the symmetry from cubic to rhombohedral and magnetic one with AF ordering into a complicated triple-k spin structure are detected in DyB_6 [6].

In terms of the $4f$ -shell filling of rare-earth ion antiferromagnets PrB_6 and NdB_6 share the places in the RB_6 sequence between heavy fermion system CeB_6 and intermediate valence compound SmB_6 . In spite of very similar paramagnetic Fermi surfaces (FS) of LaB_6 , PrB_6 and NdB_6 resulting to a common indirect RKKY-exchange motive the magnetic moments of $4f^2(\text{Pr})$ and $4f^3(\text{Nd})$ configurations are arranged into different magnetic structures. In particular, when lowering the temperature incommensurate AF phase (IC) formed in PrB_6 below Neel temperature $T_N \sim 7\text{K}$ evolves to commensurate AF one (C) observed at $T < T_M \sim 4.2\text{K}$ [7]-[9] (see Fig.1a). Application of magnetic field changes the magnetic unit cell of PrB_6 inducing the magnetic transition from C phase to another commensurate C_H phase [7]-[9] (Fig.1a). On the contrary, only one commensurate AF structure is detected in NdB_6 at $T < T_N \sim 8\text{K}$ in magnetic fields $\mu_0 H < 15\text{T}$ [10]-[12] (Fig.1b).

It should be noted here that the Fermi surface of NdB_6 in the AF state differs noticeably from that of PrB_6 . Indeed, in addition to the main FS fragment of RB_6 formed by large X-centered ellipsoids connected by the necks in X-X directions [13, 14] new regions centered at R-points are evidently established in the AF state of NdB_6 from the calculations of new specific branches detected in quantum oscillation experiments [15]. These additional folded fragments of FS induced by AF ordering could modify the strength of indirect exchange between the $4f$ -shell magnetic moments thus resulting in the different structure and parameters of the magnetic H-T phase diagrams for PrB_6 and NdB_6 (Fig.1).

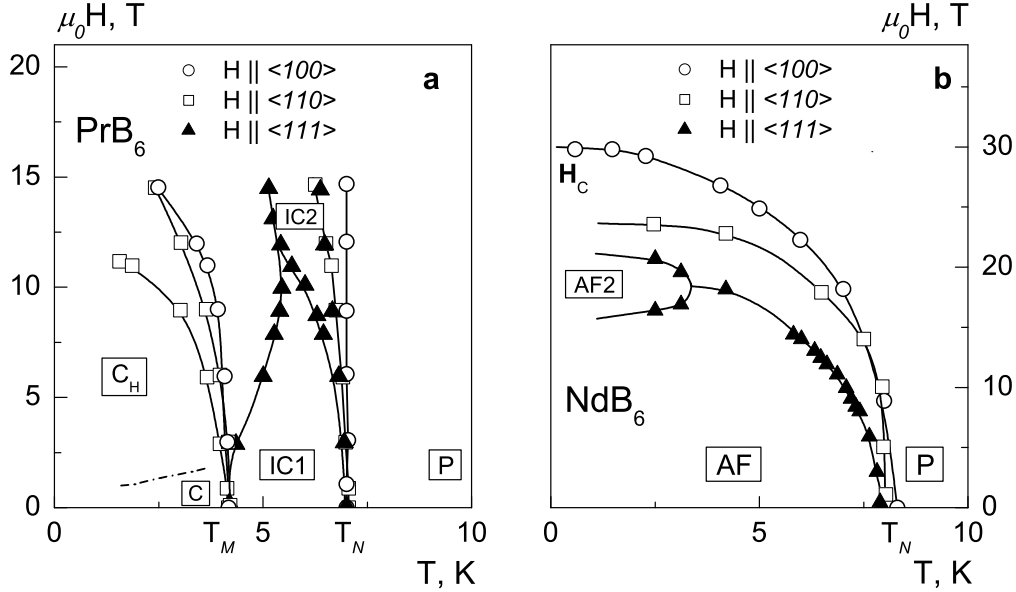


FIG. 1: H-T magnetic phase diagram of (a) PrB₆ and (b) NdB₆ for various directions of magnetic fields $\mathbf{H} \parallel \langle 100 \rangle$, $\mathbf{H} \parallel \langle 110 \rangle$, $\mathbf{H} \parallel \langle 111 \rangle$. The data are taken from [7]-[9] and [10]-[12] for PrB₆ and NdB₆ correspondingly. P, IC, C and C_H denote paramagnetic, incommensurate, commensurate and collinear magnetic phases of PrB₆. C-C_H phase transition is shown in panel (a) by dash-dotted line (see text for details).

A promising challenge to shed more light on the complicated interplay between electronic and magnetic degrees of freedom in PrB₆ and NdB₆ is provided by the study of Hall effect, which is known to be sensitive to the variation of the FS volume and topology. Available information about Hall coefficient behaviour in PrB₆ and NdB₆ given in [13, 16, 17] is fragmentary and controversial. In particular, the measurements of Hall resistivity performed in low ($\mu_0 H \approx 0.8$ T for PrB₆ and NdB₆ [13]) and moderate (up to 15 T for NdB₆ [16]) magnetic fields showed that the Hall coefficient of these compounds doesn't depend on temperature in paramagnetic state $T > T_N$. This observation contradicts evidently to the large variation of Hall resistivity (more than by a factor of 2) established in low magnetic fields $\mu_0 H = 0.1$ T and attributed to anomalous Hall effect in paramagnetic state of NdB₆ [17]. The discrepancy in experimental results and a lack of Hall effect data for strong enough magnetic fields makes it difficult to explain correctly both the exchange parameters' evolution and the magnetic

phase diagrams observed in the compounds of RB_6 family.

2. EXPERIMENTAL DETAILS

This article reports on the study of Hall effect carried out on PrB_6 and NdB_6 single crystals at temperatures $2\text{K} < T < 300\text{K}$ in magnetic fields $\mu_0 H \leq 8\text{T}$. The single crystals of rare earth hexaborides RB_6 ($\text{R}=\text{Pr}, \text{Nd}$) were grown by crucible-free inductive zone melting. X-ray diffraction and electron microprobe analysis were used to control the high quality of the grown crystals. The rectangular bar samples cut from the single crystal rods were etched in diluted nitric acid to eliminate the surface defects induced by mechanical treatment.

The angular dependencies of Hall resistivity $\rho_H(\varphi)$ have been measured by the stepwise sample rotation technique in fixed magnetic field perpendicular to rotation axis [3]. In these experiments the $\rho_H(\varphi)$ data are produced by the variation of the angle between the normal to the plane of the sample \mathbf{n} and magnetic field \mathbf{H} as a result of change in the scalar product (\mathbf{n}, \mathbf{H}) , which in turn modulates the Hall signal by harmonic law. Note that the peak-to-peak value deduced from the $\rho_H(\varphi)$ studies as the difference $\rho_H(+H) - \rho_H(-H)$ equals to this one extracted in the commonly used field sweeping technique of Hall resistivity measurements. The *dc*-current was applied along $\langle 110 \rangle$ axis taken to be parallel to the axis of rotation. High stability of magnetic field ($\Delta H/H \sim 10^{-5}$ at $\mu_0 H = 8\text{T}$) and temperature ($\Delta T \sim 0.01\text{K}$) required for this high precision measurements was achieved with the help of Cryotel SMPS-60 superconducting magnet power supply and Cryotel TC 1.5/300 temperature controller operating with LakeShore CX-1050 temperature sensor.

3. EXPERIMENTAL RESULTS

3.1. Temperature behavior of resistivity.

The temperature dependences of resistivity $\rho(T)$ measured for PrB_6 and NdB_6 in zero magnetic field are presented in Fig.2. It is seen from the data of Fig.2 that in paramagnetic state ($T > T_N$) the $\rho(T)$ curves demonstrate temperature behavior to be typical for metals. Rather high values of residual resistivity in paramagnetic phases ($\rho(10\text{K}) = 3.5\mu\Omega\cdot\text{cm}$ and $\rho(10\text{K}) = 3.9\mu\Omega\cdot\text{cm}$ for PrB_6 and NdB_6 , respectively) point to strong magnetic scattering of itinerant electrons. The onset of AF state results in a prominent decrease of resistivity down

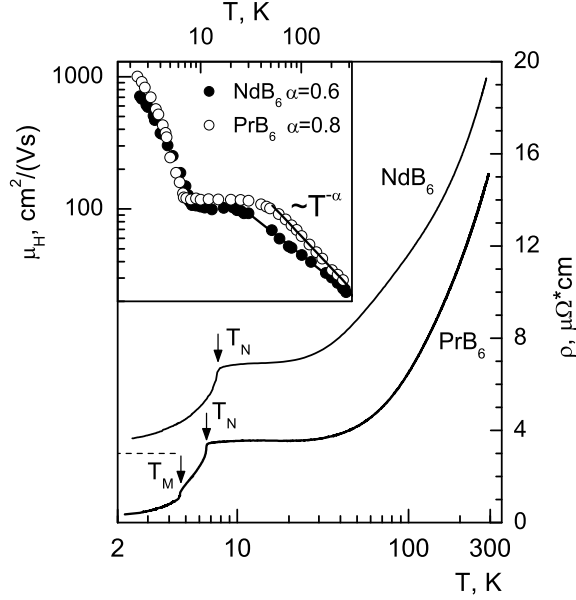


FIG. 2: Temperature behaviour of resistivity $\rho(T)$ measured for PrB_6 and NdB_6 in zero magnetic field. The $\rho(T)$ curve for NdB_6 is shifted upwards by $3\mu\Omega\cdot\text{cm}$ along vertical axis for convenience. Arrows point to magnetic phase transition temperatures. Inset shows the behaviour of Hall mobility $\mu_H(T)=R_H(T)/\rho(T)$ estimated from experimental data (see text). The solid lines in the inset represent the power law dependences $\mu_H(T)\sim T^{-\alpha}$ with the exponents given in the legend.

to the values of $0.4\mu\Omega\cdot\text{cm}$ and $0.7\mu\Omega\cdot\text{cm}$ measured at $T=2.5\text{K}$ in PrB_6 and NdB_6 , respectively. The absolute values of resistivity and the magnetic phase transition temperatures determined from $\rho(T)$ measurements ($T_N \approx 6.7\text{K}$, $T_M \approx 4.6\text{K}$ for PrB_6 and $T_N \approx 7.7\text{K}$ for NdB_6 , see Fig.2) agree with the previous data [8, 13, 16] proving the high quality of the samples under investigation.

3.2. Hall effect in PrB_6 and NdB_6

The angular dependencies of the Hall resistivity $\rho_H(\varphi)$ measured for PrB_6 and NdB_6 are presented in Fig.3a and Fig.3b, correspondingly. The angular dependencies of Hall resistivity ρ_H observed in the paramagnetic phases of these compounds could be well described by simple cosine law $\rho_H(\varphi)=\rho_{H0}+\rho_{H1}\cos\varphi$ (Fig.3, $T\geq 8\text{K}$). In vicinity of AF phase transition ($T\sim T_N$) $\rho_H(\varphi)$ curves deviate from the simple cosine behaviour and additional contribution

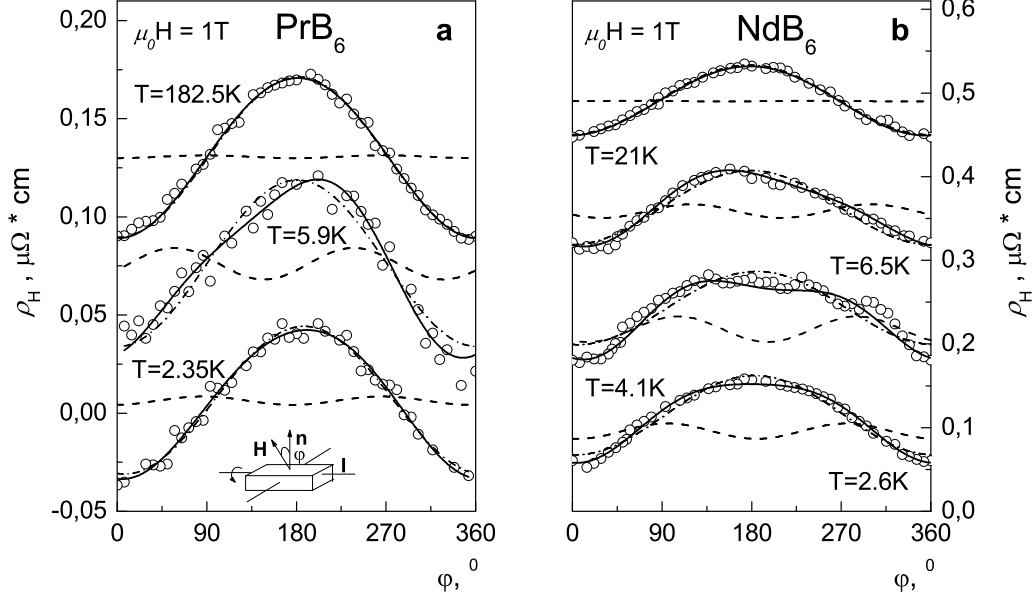


FIG. 3: Angular dependencies of Hall resistivity $\rho_H(\varphi, T_0)$ measured in magnetic field $\mu_0 H = 1\text{T}$ for (a) PrB_6 and (b) NdB_6 . The curves are shifted along a vertical axis for convenience. The lines correspond to the contributions of the first (dash-dot) and second (dash) harmonics as well as the sum of the harmonics (solid) (see Eq.1). The inset in panel (a) illustrates the experimental scheme with the sample rotation.

to the Hall effect from second harmonic $\rho_H(\varphi) \sim \cos 2\varphi$ appears in the experimental data (see, e.g., the curves $T = 5.9\text{K}$ in Fig.3a for PrB_6 and $T = 6.5\text{K}$ and 4.1K in Fig.3b for NdB_6). As a result, the $\rho_H(\varphi)$ dependences may be fitted by relation

$$\rho_H(\varphi) = \rho_{H0} + \rho_{H1} \cos(\varphi) + \rho_{H2} \cos(2\varphi - \Delta\varphi), \quad (1)$$

where ρ_{H0} is a constant value arising due to the misalignment of Hall probes, ρ_{H1} and ρ_{H2} are the amplitudes of the main and second harmonic and $\Delta\varphi$ is the phase shift of the second harmonic.

Note that the transverse configuration of the Hall experiment used in our study (the rotational axis of the sample is parallel to vector \mathbf{I} and perpendicular to vector \mathbf{H} ; see the inset in Fig.3a) allows to minimize the spurious magnetoresistance contribution to the Hall signal, which could also result in even harmonic [3, 18]. To estimate independently the possible contribution to Hall effect induced by anisotropic magnetoresistance $\sim \cos(2\varphi)$, the

angular dependences of Hall resistance and magnetoresistance were recorded simultaneously with subsequent scaling of magnetoresistance data $\rho(\varphi)$ to ρ_{H0} emerging due to misalignment of Hall probes. Our estimations lead to conclusion that the magnetoresistive component has no appreciable effect on Hall resistivity $\rho_H(\varphi)$ in magnetic fields up to 8T.

The approach described above was earlier applied to separate the various contributions to Hall effect in the AF phases of antiferromagnet CeAl_2 [18] and heavy fermion compound CeB_6 [3]. Besides, similar technique was successfully used to establish and explain the complicated behavior of Hall coefficient in rare earth dodecaborides RB_{12} [19, 20] and in metallic systems with heavy fermions CeAl_3 [21] and quantum critical behavior $\text{CeCu}_{6-x}\text{Au}_x$ [22].

In present study the $\rho_H(\varphi)$ data were fitted by (Eq.1) both in paramagnetic and AF phases of RB_6 (R=Pr, Nd). As a result, the temperature and magnetic field behaviour of the Hall resistivity $\rho_{H1}(\text{T}, \text{H})$, even harmonic term $\rho_{H2}(\text{T}, \text{H})$ and the phase shift $\Delta\varphi(\text{T}, \text{H})$ to be deduced from the experimental data are presented and discussed in the next sections.

3.3. Temperature and magnetic field dependences of Hall coefficient in PrB_6 and NdB_6

The amplitude of the first harmonic term ρ_{H1} was used to calculate the Hall coefficient $R_H(\text{T}) = \rho_{H1}(\text{T})/H$ (d is the sample thickness). The temperature dependencies of $R_H(\text{T})$ obtained for PrB_6 and NdB_6 are presented in Fig.4a and Fig.4b, correspondingly. The $R_H(\text{T})$ data for nonmagnetic reference compound LaB_6 ($4f^0$ configuration) is also shown in Fig.4a for comparison. It is found that Hall coefficient increases for both PrB_6 and LaB_6 above liquid nitrogen temperature (Fig.4a), but for NdB_6 it decreases only slightly when temperature rises in the interval $70\text{K} \leq \text{T} \leq 300\text{K}$ (Fig.4b). At the same time, the data in Fig.4 demonstrate that Hall coefficient measured in magnetic field $\mu_0 H = 1\text{T}$ for PrB_6 and NdB_6 doesn't depend noticeably on temperature in the range of $8 \div 70\text{K}$. Note also that in this temperature interval the estimated values of Hall coefficient $R_H(\text{PrB}_6) \approx -(4.2 \pm 0.1) \cdot 10^{-4} \text{ cm}^3/\text{C}$ and $R_H(\text{NdB}_6) \approx -(4.1 \pm 0.1) \cdot 10^{-4} \text{ cm}^3/\text{C}$ agree with the data of Onuki et al [13] (see also Fig.5a).

The temperature dependence of the $R_H(\text{T}, \mu_0 H = 1\text{T})$ in the paramagnetic phase of NdB_6 (Fig.4b) contradicts to the low field Hall effect data obtained in [17]. In contrast to

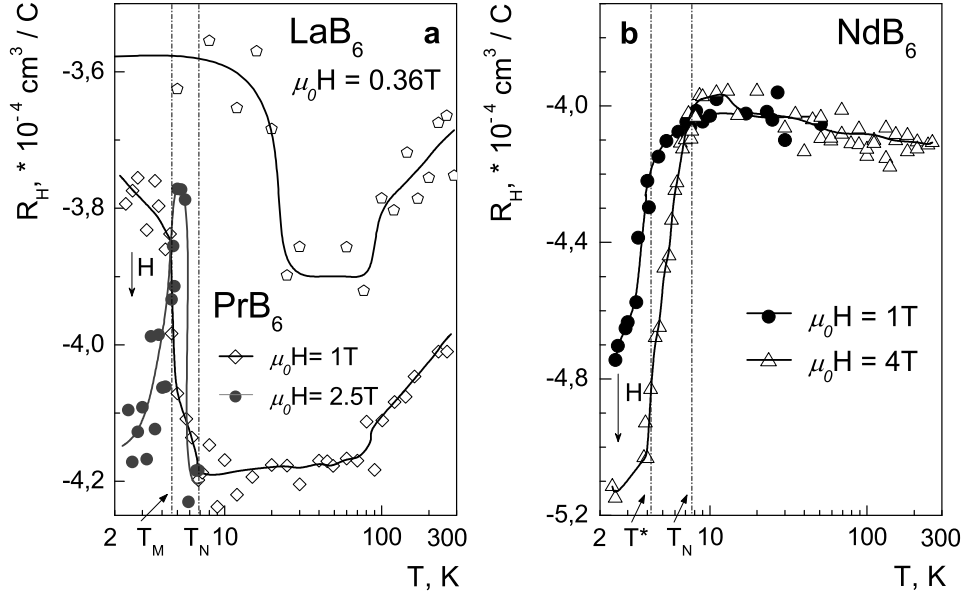


FIG. 4: Temperature dependences of Hall coefficient $R_H(T)$ for (a) LaB_6 , PrB_6 and (b) NdB_6 in magnetic fields $\mu_0 H \leq 4\text{T}$. Solid lines are drawn to guide for eye.

the present work, Hall resistivity measured for the same current direction $\mathbf{I} \parallel \langle 110 \rangle$ in [17] demonstrates a pronounced peak of $\rho_H(T)$ at $T_N \sim 8\text{K}$, which is followed by a gradual diminishing of R_H when the temperature increases up to room value (Fig.5a). To verify this issue the magnetic field dependences of Hall resistivity $\rho_H(H)$ were measured for NdB_6 at temperatures below and above T_N in magnetic fields up to 8T. Special attention was paid to the range of low magnetic fields $\mu_0 H \leq 1\text{T}$, where drastic difference of $R_H(T)$ values measured in [13, 17] is observed (to compare $R_H(10\text{ K}) = -(4.6 \pm 0.9) \cdot 10^{-4} \text{ cm}^3/\text{C}$ [13] and $R_H(10\text{ K}) = \rho_H(10\text{ K}) - 4 \cdot 10^{-4} \text{ cm}^3/\text{C}$ [17], see Fig.5a). The comparison between the $\rho_H(H)$ data obtained in the present study and in [17] (Fig.5b) shows that Hall resistivity depends linearly on magnetic field demonstrating no contribution of second harmonic in the considered range of temperatures and magnetic fields. As a result, the data of Fig.5 allow to conclude that in the paramagnetic state of NdB_6 Hall coefficient doesn't depend noticeably on temperature and magnetic field showing only small (less than 10%) variation of R_H (Fig.5a). Therefore our data do not confirm earlier observed Hall coefficient anomaly at $T \sim T_N$ [17] (Fig.5a) and it makes questionable also the interpretation in terms of the

anomalous Hall effect in the paramagnetic phase of NdB₆ proposed in [17].

The transition to AF state is accompanied by a different variation of the low field Hall coefficient in PrB₆ and NdB₆. For $T < T_N$ the absolute value of $R_H(T)$ in PrB₆ decreases drastically ($\Delta R_H/R_H \sim 10\%$) when temperature is lowered (see curve $\mu_0 H = 1\text{T}$ in Fig.4a). At the same time, the absolute value of $R_H(T)$ in NdB₆ increases noticeably ($\sim 15\%$ for $\mu_0 H = 1\text{T}$) just below the Neel temperature (Fig.4b). In our opinion, this difference in $R_H(T)$ behaviour may be attributed to the peculiarities of AF phases and it will be discussed in the last section.

3.4. Second harmonic contribution in Hall effect

In paramagnetic phase of PrB₆ and NdB₆ the contribution of ρ_{H2} is very small as compared to the first harmonic. However, the amplitude ρ_{H2} rises drastically when AF state sets up in PrB₆ and NdB₆ (Fig.6). It is worth noting that for PrB₆ the term ρ_{H2} contributes essentially to Hall signal in low fields $\mu_0 H \leq 1\text{T}$ only in the incommensurate AF phase and the component was evidently observed in the range $T_M < T < T_N$ (see Fig.1a), while for NdB₆ the temperature dependence of $\rho_{H2}(T)$ is characterized by a pronounced maximum at intermediate temperature $T^* \sim 4\text{K}$, which is well below T_N (Fig.6). The elevation of magnetic field results in an essentially different behaviour of the second harmonic term in PrB₆ and NdB₆. Indeed, a pronounced increase of the $\rho_{H2}(T)$ values in PrB₆ is accompanied by broadening of the peak in magnetic field (Fig.6a). On the contrary, for NdB₆ the $\rho_{H2}(T)$ maximum value decreases evidently for $\mu_0 H > 1\text{T}$ (see, e.g., data for $\mu_0 H = 4\text{T}$ in Fig.6b).

Interesting that noticeable anomalies of both magnetoresistance [23, 24] and C_{44} elastic constant [25] temperature dependencies have been earlier detected in the vicinity of temperature T^* for NdB₆. These features at $T^* \sim 4\text{K}$ may indicate on the possible changes of electronic and/or magnetic structure occurred in the commensurate AF phase of NdB₆ well below the Neel temperature. However, a detailed investigation of magnetic and charge transport parameters of NdB₆ need to be carried out to shed more light on the origin of the Hall effect anomalies observed at T^* in AF phase of NdB₆.

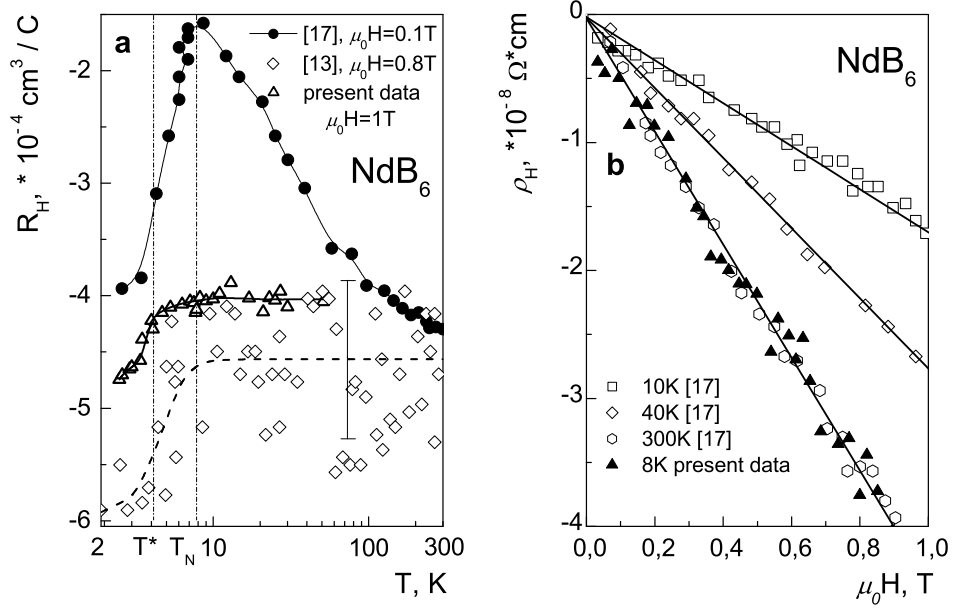


FIG. 5: (a) The comparison of $R_H(T)$ temperature dependences obtained for NdB_6 in this study with those ones reported in [13] and [17]. Panel (b) presents the field dependences of low field ($\mu_0 H \leq 1 \text{ T}$) Hall resistivity $\rho_H(H)$ in comparison with the data of [17].

4. DISCUSSION

4.1. Paramagnetic phase of PrB_6 and NdB_6 ($T > T_N$)

High accuracy of data obtained allows us to estimate the number of conduction electrons per unit cell in the paramagnetic phase of rare earth hexaborides under investigation. The values $n/n_{4f}(\text{PrB}_6) \approx 1.05 \pm 0.01$ and $n/n_{4f}(\text{NdB}_6) \approx 1.09 \pm 0.01$, which are established just above the Neel temperature (at 10 K) in these magnetic hexaborides, are comparable with charge carriers' concentration previously estimated for CeB_6 ($n/n_{4f} \approx 1.00$ [3]). An approximately linear increase of n/n_{4f} parameter vs $4f$ -shell occupation in the magnetic hexaborides agrees well with the results obtained in [26, 27], where a remarkable expansion of the small FS electron pockets was detected in the light RB_6 compounds. The increase of n/n_{4f} produced by FS changes is a factor, which is responsible for a variation of both RKKY-function $\Sigma F(2k_F R_i)$ (here k_F and R_i denote Fermi wavevector and the distance between the magnetic moments of the rare earth ions, respectively) and indirect exchange interaction.

According to de Gennes approach [28], the Neel temperature T_N , reduced concentration n/n_{4f} , exchange constant J_{ex} , Fermi energy E_F , de Gennes factor $G=(g-1)^2J(J+1)$ and RKKY-function $\Sigma F(2k_F R_i)$ are related by the expression

$$T_N = \frac{3\pi}{4} \left(\frac{n}{n_{4f}} \right)^2 G \left(\frac{J_{ex}^2}{E_F} \right) \Sigma F(2k_F R_i). \quad (2)$$

Taking into account both the strong variation of the de Gennes factor ($G(\text{PrB}_6) \approx 0.8$ and $G(\text{NdB}_6) \approx 1.84$) and 5% increase in the charge carriers concentration n/n_{4f} together with only small changes in the lattice constant and, hence, in the J_{ex} exchange parameter, one needs to propose an essential decrease of the $\Sigma F(2k_F R_i)$ values from PrB_6 to NdB_6 , which is necessary to explain relatively small changes in Neel temperatures from $T_N(\text{PrB}_6) \approx 6.7\text{K}$ to $T_N(\text{NdB}_6) \approx 7.7\text{K}$. Indeed, the pronounced lowering of RKKY-function was predicted for RB_6 compounds when the n/n_{4f} ratio increases in the range $1 - 1.2$ [29], but more detailed calculations are necessary to estimate quantitatively the variation of T_N in the rare earth hexaborides family.

4.2. Antiferromagnetic state of PrB_6 and NdB_6 ($T < T_N$)

When discussing the complicated behavior of Hall effect in AF phases of PrB_6 and NdB_6 (see sections 3.3 – 3.4) it is worth to mention that the magnetic structures of PrB_6 and NdB_6 have different parameters in low magnetic field region. The magnetic unit cell of PrB_6 C-phase involves 32 structural unit cells [30] and characterizes by a wave vector $\mathbf{Q} = (1/4, 1/4, 1/2)$ while simple doubling of the structural unit cell is observed in NdB_6 [31]. On the contrary, the structure of PrB_6 commensurate C_H magnetic phase formed in moderate magnetic fields (Fig.1a) is similar to that one of commensurate AF phase observed in NdB_6 in magnetic fields below 15T [7, 9, 10]. In this respect the dramatic change of Hall coefficient R_H found in PrB_6 with the increase of magnetic field from 1T to 2.5T (Fig.4a) may be definitely associated with the crossing of the C- C_H phase boundary below T_M .

To obtain more information about the variation of Hall coefficient when entering from C to C_H phases of PrB_6 , the magnetic field dependencies of $R_H(H, T_0)$ have been obtained from the experimental data for fixed temperatures $T_0 < T_M$ in the range $\mu_0 H \leq 6\text{T}$. The $R_H(H, T_0)$ data shown in Fig.7a allow to detect clearly the C- C_H phase transition in PrB_6 establishing a positive slope of C- C_H phase boundary in agreement with the results [8, 9]

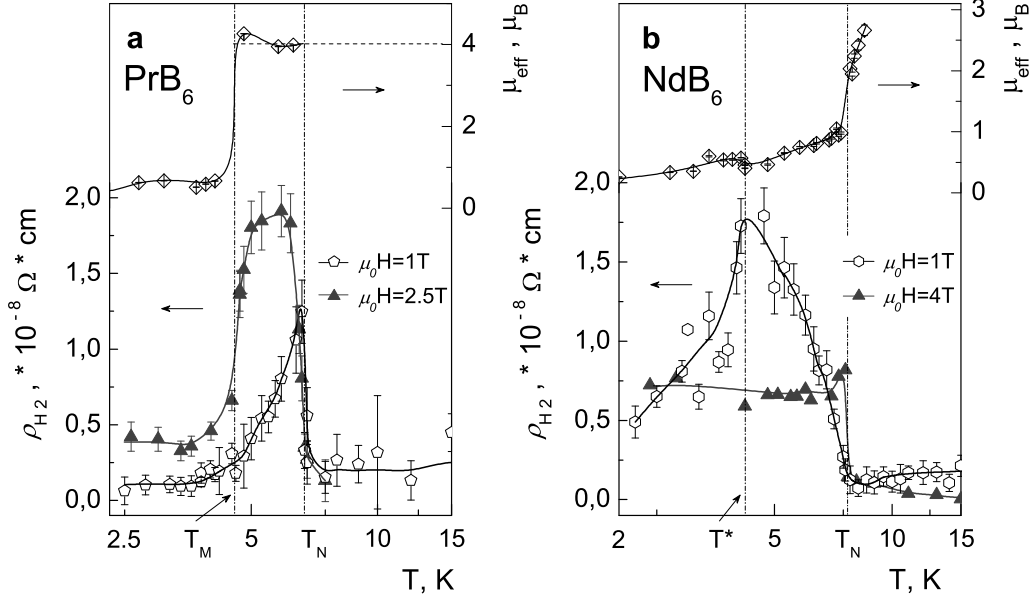


FIG. 6: Temperature dependencies of the amplitude of the second harmonic term ρ_{H2} measured for (a) PrB_6 and (b) NdB_6 in magnetic fields $\mu_0 H \leq 4\text{T}$. The upper curves on the both panels represent the temperature dependencies of the effective magnetic moment μ_{eff} [24].

(see also Fig.1a). So, the similar behaviour of the Hall coefficient temperature dependencies for PrB_6 at $T < T_M$ (see data for $\mu_0 H = 2.5\text{T}$ in Fig.5a) and for NdB_6 at $T < T_N$ (Fig.5b) may be likely understood assuming identical magnetic structures (simple type I antiferromagnet with ordering vector $\mathbf{Q} = (0, 0, 1/2)$) developing in the AF phase of NdB_6 (Fig.1b) and in the C_H -phase of PrB_6 (Fig.1a).

The strong renormalization of the Hall coefficient $R_H(T)$ in the commensurate phases of the studied antiferromagnets (Figs.5,7) may be attributed to the reconstruction of FS below the AF phase transition. The electronic structure of the AF state in these hexaborides may be properly understood through folding of the paramagnetic band structure. Following to the FS reconstruction of [15], the simple cubic Brillouin zone in the paramagnetic state of RB_6 is reduced by this folding procedure to the tetragonal one in the AF phase of NdB_6 . As a result, two kinds of electron sheets and one hole sheet appear in the AF phase of NdB_6 causing to the remarkable changes in the Hall coefficient behaviour with temperature both in weak and strong magnetic field regimes. However, the independent verification of this

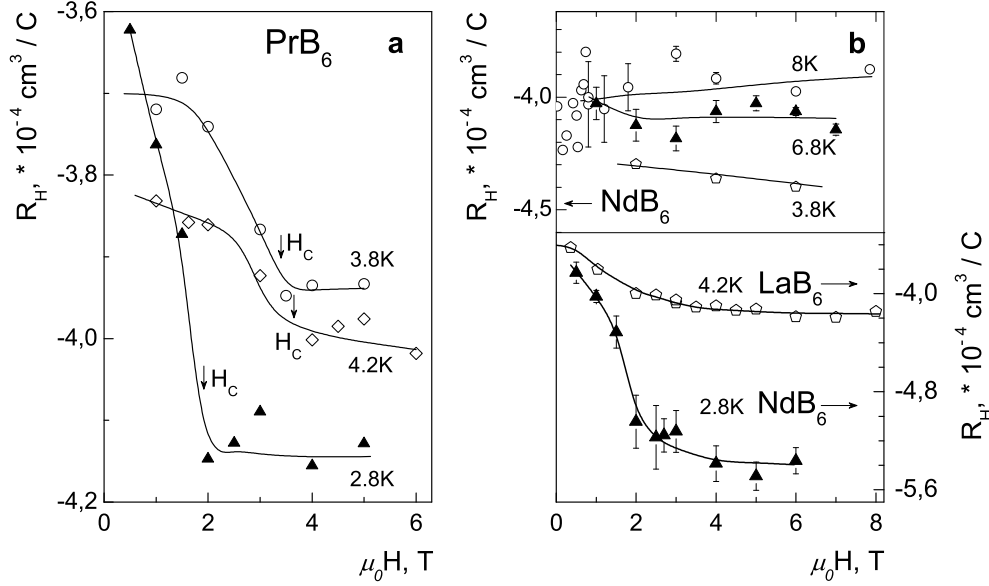


FIG. 7: The field dependencies of Hall coefficient $R_H(H)$ for (a) PrB_6 and (b) NdB_6 in the temperature region $T \leq 8\text{K}$. Arrows in panel (a) point to the characteristic fields of C- C_H transition. The data of LaB_6 are given for comparison in panel (b).

scenario requires a comparative study of Hall effect and magnetization to be carried out in strong enough magnetic fields.

The resistivity (Fig.2) and Hall effect (Fig.4) data were used to estimate Hall mobility $\mu_H(T) = |R_H(T)|/\rho(T)$ for PrB_6 and NdB_6 (inset in Fig.2). In commensurate AF phases of these compounds Hall mobility gets values $\mu_H(\text{PrB}_6) \approx 1000 \text{ cm}^2/(\text{V}\cdot\text{s})$ and $\mu_H(\text{NdB}_6) \approx 710 \text{ cm}^2/(\text{V}\cdot\text{s})$. In paramagnetic state in the range $45\text{K} \leq T \leq 300\text{K}$ the $\mu_H(T)$ curves could be well fitted by the power law dependence $\mu_H \sim T^{-\alpha}$ with the exponents $\alpha(\text{PrB}_6) \sim 0.8$ and $\alpha(\text{NdB}_6) \sim 0.6$ (inset in Fig.2). The observed decrease of both Hall mobility μ_H and exponent α when moving from PrB_6 to NdB_6 may be associated with the enhancement of magnetic scattering of itinerant electrons on the localized magnetic moments of $4f$ -states of R^{3+} ions ($\text{R}=\text{Pr}, \text{Nd}$) that agrees well with de Gennes scaling rule [28].

Finally, it is worth to note a very large difference in the low temperature values of Hall mobility μ_H estimated from the experimental data for non-magnetic reference compound LaB_6 ($\mu_H(4.2\text{K}) \approx 18000 \text{ cm}^2/(\text{V}\cdot\text{s})$, $\rho(4.2\text{K}) \approx 0.016 \mu\Omega\cdot\text{cm}$) and antiferromagnetic PrB_6

and NdB_6 ($\mu_H(4.2\text{K}) \approx 370 - 430 \text{ cm}^2/(\text{V}\cdot\text{s})$, see inset in Fig.2). As a result, the remarkable changes of the Hall coefficient in magnetic field observed in the present study for LaB_6 and NdB_6 (Fig. 7b) should be explained by quite different factors. In the case of nonmagnetic LaB_6 the strong enough variation of $R_H(H, 4.2\text{K})$ (by $\sim 16\%$) could be certainly attributed to transition from weak ($\omega\tau \ll 1$, where ω and τ are cyclotron frequency and charge carriers' relaxation time, correspondingly) to strong ($\omega\tau \gg 1$) magnetic field regime [32]. However, rather low mobility of charge carriers in NdB_6 doesn't allow to interpret the drastic changes of $R_H(H)$ observed in the AF phase of NdB_6 (see, e.g., curve for $T = 2.8\text{K}$ in Fig.7b) in terms of the $\omega\tau$ approach.

To explain the anomalous behaviour of Hall coefficient in the studied rare earth hexaborides it is necessary to address to the results of transverse magnetoresistance study performed recently for these compounds [24]. In particular, it was shown [24] that nanoscale magnetic clusters with strongly renormalized effective magnetic moments $\mu_{eff}(\text{PrB}_6) \approx 4\mu_B$ and $\mu_{eff}(\text{NdB}_6) \approx 2.5\mu_B$ are formed just above the Neel temperature in these compounds (see also $\mu_{eff}(T)$ curves in Fig.6). The unit cell magnetic clusters' formation attributed to the exchange induced $5d$ -states' spin polarization [24] was proposed to be responsible both for the effects of density-of-states renormalization and the formation of additional $5d$ -component in the magnetic structure of these unusual antiferromagnets. In such a case the interaction between the spin polarized component of $5d$ -states and the magnetic structure of $4f$ magnetic moments in the rare earth hexaborides under investigation could be considered as an important reason resulting both in the appearance of the magnetic anisotropy in AF-phase [23, 24] and in the noticeable renormalization of Hall effect observed just below T_N in RB_6 under investigation (Fig.4).

5. CONCLUSION

To summarize, the Hall effect in the antiferromagnetic metals PrB_6 and NdB_6 has been studied at temperatures in the range $2 - 300\text{K}$ in magnetic fields up to 8T . The detailed comparison between the $R_H(T)$ temperature dependencies obtained in present investigation and those ones reported earlier in [13, 17] allows establishing temperature independent behaviour of $R_H(T)$ in paramagnetic state of these two RB_6 compounds excluding the interpretation [17] in terms of anomalous paramagnetic Hall effect in NdB_6 . Within the analysis based

on the de Gennes approach an essential decrease of RKKY-function amplitude from PrB_6 to NdB_6 is suggested to be the main reason of the close magnitudes of Neel temperatures in these magnetic hexaborides with very different values of the de Gennes factors. Additionally, quite different behaviour of $R_H(T)$ was found below T_N for PrB_6 and NdB_6 in low magnetic fields. It was shown that the transition to the commensurate C_H phase in PrB_6 is accompanied by the pronounced (up to 10%) decrease of Hall coefficient R_H at liquid helium temperatures. As a result the temperature behaviour of $R_H(T)$ in the C_H phase of PrB_6 is proved to be similar to that one found in the commensurate AF phase of NdB_6 . Our findings favour the enhancement of the magnetic scattering of the charge carriers on the localized magnetic moments of R^{3+} ions when moving from PrB_6 to NdB_6 . The observed variation of Hall coefficient in the AF phases of these RB_6 compounds is supposed to be induced by the effects of paramagnetic FS structure folding and the effects of density-of-states renormalization, which could be attributed to the magnetic polarization of $5d$ -states both in AF and paramagnetic states of the hexaborides.

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- [1] Sluchanko N E, Glushkov V V, Demishev S V, Samarin N A, Bogach A V, Gon'kov K V, Khayrullin E I, Filipov V B, Shitsevalova N Yu *Physica B: Cond. Mat.* **403**, 1393 (2008)
 - [2] Takada K, Shintake T, Baba H, Inagaki T, Onoe K, Tanaka T, Matsumoto H *Proceedings of the FEL Conference* 351 (2004)
 - [3] Sluchanko N E, Bogach A V, Glushkov V V, Demishev S V, Ivanov V Yu, Ignatov M I, Kuznetsov A V, Samarin N A, Semeno A V *JETP* **104**, 120(2007)
 - [4] Wachter P *Handbook on the Physics and Chemistry of Rare Earths* **19** eds. Gschneidner K A Jr., Eyring L, Lander G H, Choppin G R Elsevier Science B.V. 177-382 (1994)
 - [5] Süllo S, Prasad I, Aronson M C, Bogdanovich S, Sarrao J L, and Fisk Z *Phys. Rev. B.* **62**,

11626 (2000)

- [6] Takahashi K, Nojiri H, Ohoyama K, Ohashi M, Yamaguchi Y, Kunii S, Motokawa M *Physica B* **241-243**, 696 (1998)
- [7] Sera M, Kim M S, Tou H, Kunii S *J. Phys. Soc. Jpn.* **73**, 3422 (2004)
- [8] Kobayashi S, Sera M, Hiroi M, Nishizaki T, Kobayashi N, Kunii S *J. Phys. Soc. Jpn.* **70**, 1721 (2001)
- [9] Iwakubo H, Ikeda S, Kishino Y, Tanida H, Sera M, and Iga F *Phys. Rev. B* **78**, 012409 (2008)
- [10] Awaji S, Kobayashi N, Sakatsume S, Kunii S and Sera M *J. Phys. Soc. Jpn.* **68**, 2518 (1999)
- [11] Goodrich R G, Harrison N, and Fisk Z *Phys. Rev. Lett.* **97**, 146404 (2006)
- [12] Yonemura T, Tanida H, Sera M, and Iga F *J. Phys. Soc. Jpn.* **78**, 114705 (2009)
- [13] Onuki Y, Umezawa A, Kwok W K, Grabtree G W, Nishihara M, Yamazaki T, Omi T and Komatsubara T *Phys. Rev. B* **40**, 11195 (1989)
- [14] Endo M., Nakamura S., Isshiki T., Kimura N., Nojima T., Aoki H., Harima H., Kunii S. *J. Phys. Soc. Jpn.* **75**, 114704 (2006)
- [15] Kubo Y, Asano H, Harima H and Yanase A *J. Phys. Soc. Jpn.* **62**, 202 (1993)
- [16] Sera M, Hiroi M, Kobayashi N, Kunii S *J. Phys. Soc. Jpn.* **67**, 629 (1998)
- [17] Stankiewicz Y, Nakatsuji S, Fisk Z *Phys. Rev. B* **71** 134426 (2005)
- [18] N. E. Sluchanko, A. V. Bogach, V. V. Glushkov, S. V. Demishev, M. I. Ignatov, N. A. Samarin, G. S. Burkhanov, and O. D. Chistyakov, *JETP* **98**, 793 (2004)
- [19] N. Sluchanko, L. Bogomolov, V. Glushkov, S. Demishev, M. Ignatov, Eu. Khayrullin, N. Samarin, D. Sluchanko, A. Levchenko, N. Shitsevalova and K. Flachbart, *Phys. Status Solidi B* **243**, R63 (2006)
- [20] N. E. Sluchanko, D. N. Sluchanko, V. V. Glushkov, S. V. Demishev, N. A. Samarin, and N. Yu. Shitsevalova, *JETP Lett.* **86**, 604 (2007)
- [21] N. E. Sluchanko, A. V. Bogach, G. S. Burkhanov, O. D. Chistyakov, V. V. Glushkov, S. V. Demishev, N. A. Samarin, and D. N. Sluchanko, *Physica B* **359-361**, 308 (2005)
- [22] N. E. Sluchanko, D. N. Sluchanko, N. A. Samarin, V. V. Glushkov, S. V. Demishev, A. V. Kuznetsov, G. S. Burkhanov and O. D. Chistyakov *Low Temp. Phys.* **35**, 544 (2009)
- [23] Anisimov M, Bogach A, Glushkov V, Demishev S, Samarin N, Filipov V, Shitsevalova N, Sluchanko N *J.Phys.:Conf.Ser.* **200**, 032003 (2010)
- [24] Anisimov M A, Bogach A V, Glushkov V V, Demishev S V, Samarin N A, Filipov A B,

- Shitsevalova N Yu, Sluchanko N E *JETP* **109**, 815 (2009)
- [25] Nakamura S, Goto T, Kunii S, Iwashita K, Tamaki A *J. Phys. Soc. Jpn.* **63**, 623 (1994)
- [26] Onuki Y, Komatsubara T, Reinders P H P, Springford M *J. Phys. Soc. Jpn.* **58**, 3698 (1989)
- [27] Behler S, Winzer K *Z. Phys B* **82**, 355 (1991)
- [28] de Gennes P G *J. Phys. Radiat.* **23**, 510 (1962)
- [29] Baranovskiy A E, Grechnev G E, Fil V D, Ignatova T V, Logosha A V, Panfilov A S, Svechkarev I V, Shitsevalova N Yu, Filippov V B, Eriksson O *J. Alloys and Comp.* **442**, 228 (2007)
- [30] C. M. McCarthy, C. W. Thompson, R. J. Graves, H. W. White, Z. Fisk, and H. R. Ott, *Solid State Commun.* **36**, 861 (1980)
- [31] C. M. McCarthy and C. W. Thompson, *J. Phys. Chem. Solids* **41**, 1319 (1980)
- [32] I. M. Lifshitz, M.Ya.Azbel and M. I. Kaganov. *Electron Theory of Metals*. Moscow, Nauka (1971). Translated: NewYork: Consultants Bureau (1973)